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Tritium Concentrations in Vegetation as a Function of Distance from a Low-Level Radioactive Waste Site at Los Alamos National Laboratory



Edited by Hector Hinojosa, Group IM-1 Prepared by Teresa Hiteman, Group RRES-ECO

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L. M. Vasquez-Tator

E. A. Lopez



# TRITIUM CONCENTRATIONS IN VEGETATION AS A FUNCTION OF DISTANCE FROM A LOW-LEVEL WASTE SITE AT LOS ALAMOS NATIONAL LABORATORY

P.R. Fresquez, L.M. Vasquez-Tator, and E.A. Lopez

#### **ABSTRACT**

Low-level radioactive wastes have been buried at Area G at Los Alamos National Laboratory since 1957. Wastes are buried in pits, trenches, or shafts and then covered with fill material. Tritium (<sup>3</sup>H), a radioactive isotope of hydrogen, is one of the main contaminants in waste materials at Area G and has been consistently detected in annual environmental monitoring studies of soils and plants. The purpose of this study was to determine the extent of <sup>3</sup>H migration from the perimeter of Area G using vegetation as an investigative tool. Piñon and juniper trees, in particular, are excellent indicators of subterranean <sup>3</sup>H migration. Tree branch tips, collected at chest height, were collected at various distances (approximately 10, 50, 100, 150, and 200 m) from the perimeter of Area G in seven directions (N, NE, E, SE, S, SW, and NW). Most <sup>3</sup>H concentrations in trees collected nearest the perimeter boundary (10 to 16 m) around Area G were detectable (where the result was higher than the analytical uncertainty at the 99% confidence level) and higher than the regional statistical reference level (>0.50 pCi/mL). From there, most transects (NE, E, S, and NW) showed decreasing concentrations with distance; two remained constant (SE and SW), and one transect (N) showed a quadratic response. The highest <sup>3</sup>H concentration (6,700 pCi/mL) was detected on the south side of Area G nearest the <sup>3</sup>H shafts at the 10- to 16-m distance; this amount, however, was far below the toxicity reference value.

#### I. INTRODUCTION

Radioactive wastes have been disposed of by burial at Los Alamos National Laboratory (LANL) since the early 1940s (Purtymun et al., 1980). Area G is a 25.5-hectare (63-acre), low-level radioactive waste processing and disposal area located on the east end of Mesa del Buey at Technical Area (TA) 54 (Figure 1). Area G was established in 1957 and is the Laboratory's primary radioactive-solid-waste burial and storage site (Soholt 1990). Wastes for disposal include contaminated equipment, paper, plastics, clothing, building materials, soils, and process wastes and are placed in either pits, trenches, or shafts and then covered with fill material (Hansen et al., 1980). Tritium (<sup>3</sup>H),

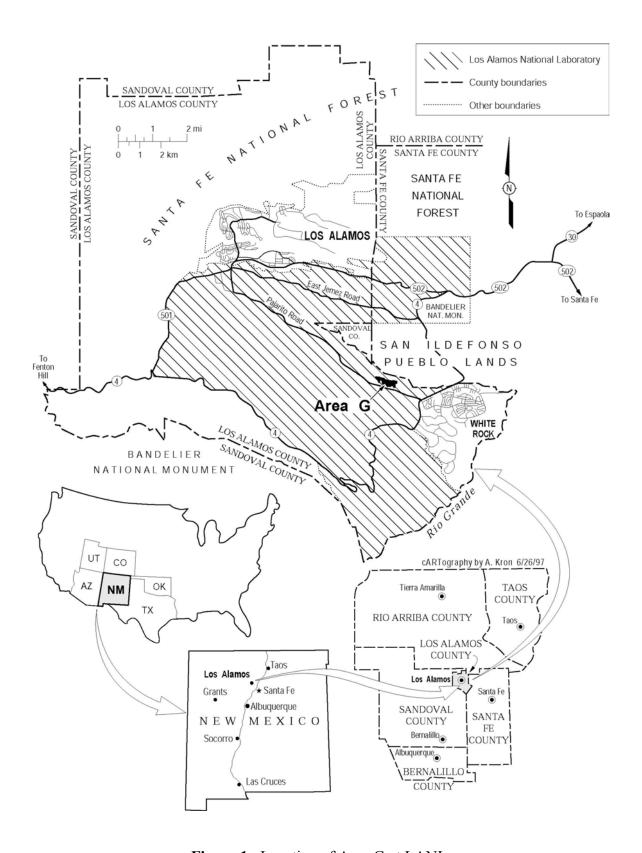


Figure 1. Location of Area G at LANL.

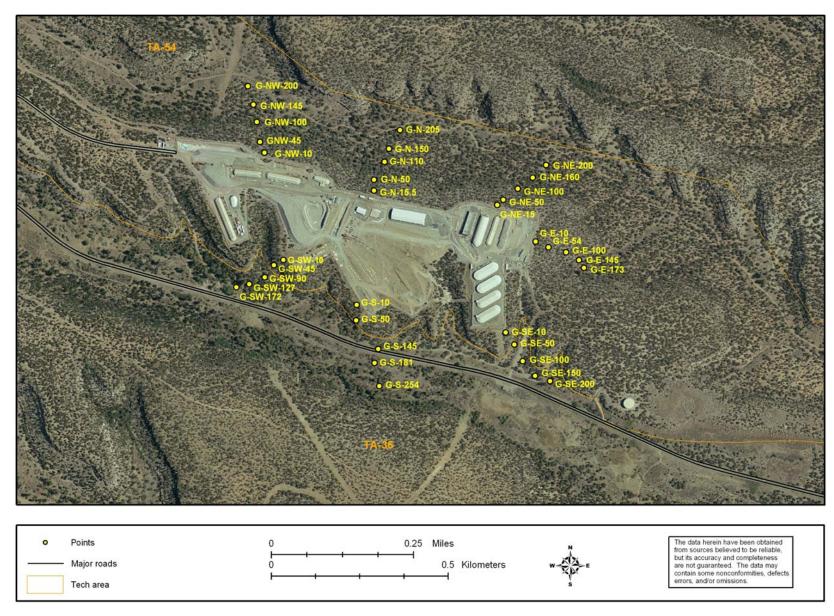
uranium, plutonium, and a variety of fission and activation products are the main isotopes in waste materials deposited at Area G (US DOE 1979).

As part of the Environmental Surveillance Program at LANL, soil and vegetation are collected annually from within and around the Area G facility to help monitor and assess the site's impact on the surrounding environment (Lopez 2002). Of all the radionuclides that have been assessed, plutonium and <sup>3</sup>H are routinely detected above regional background concentrations (Jacobson 1992; Fresquez et al., 1995; Mullen et al., 1996; Conrad et al., 1996; Nyhan et al., 2001). Tritium, in particular, has been shown to be leaking from underground storage shafts (Purtymun 1973) and has exceeded screening action levels in soils (Nyhan et al., in preparation). Also, monitoring results have shown that <sup>3</sup>H has migrated beyond the fence perimeter.

The uptake of radionuclides by vegetation may give some insight into surface (Hansen et al., 1980) and subsurface (Wenzel et al., 1987) pathways of contaminants to humans from waste disposal areas. Trees, in particular, have been shown to be excellent indicators of subterranean <sup>3</sup>H migration from low-level radioactive waste disposal sites (Rickard and Kirby 1987). This paper reports the extent of <sup>3</sup>H migration from the perimeter boundary fence of Area G via vegetation monitoring.

#### II. METHODS AND MATERIALS

Seven transects were established to the north, northeast, east, southeast, south, southwest, and northwest of Area G (Figure 2). Along each transect, samples were collected at approximately 10, 50, 100, 150, and 200 m according to availability of vegetation and landscape. Actual distances of each transect can be found in Figure 2 and Appendix A. At each sampling point, approximately 1.4 kg (3 lb) of tree clippings were collected with clean sheers at chest height and placed into Ziplock bags. The Ziplock bags were then placed into a cooler containing ice and transported back to the laboratory. At the laboratory, the (unwashed) samples were placed into a distillation unit and heated to collect the distillate moisture. The water samples (about 20 mL) were placed into labeled 50-mL plastic vials, taped with chain-of-custody tape, and shipped to Paragon Analytics, Inc., Fort Collins, Colorado, for analysis. All of the original data can be found in Appendix B for reference.



**Figure 2.** Locations of transects and sample points from Area G at TA-54.

#### III. RESULTS

In general, most vegetation samples collected nearest the fence perimeter contained <sup>3</sup>H concentrations that were detectable (where the result is higher than the total propagated analytical uncertainty [TPU] at the 99% confidence level) and higher than the Regional Statistical Reference Level (RSRL) of 0.50 pCi/mL and then generally decrease with distance in most transects (Table 1 and Figures 3 and 4). The TPU is the measure of all known sources of potential error (Nyhan et al., 2002); and a detectable concentration was considered to be a result that was greater than three times the TPU (Corley et al., 1981). The RSRL is the upper level background concentration (mean plus two standard deviations = 95% confidence level), based on data collected from regional background locations during the years 1998 to 2002 (Nyhan et al., in preparation). The results by transect are the following:

- North Transect: All vegetation samples collected along this transect contained detectable <sup>3</sup>H concentrations higher than the RSRL. In fact, the concentrations of <sup>3</sup>H with distance showed a quadratic response with the highest concentrations of <sup>3</sup>H nearest the fence perimeter and at the 205-m mark.
- Northeast Transect: There were significant concentrations of <sup>3</sup>H at the 10- to 16- m mark and at the 45- to 54-m mark; the concentrations of <sup>3</sup>H then generally decreased thereafter to the RSRL.
- East Transect: The 10- to 16-m sample contained <sup>3</sup>H at concentrations just above the RSRL. All other samples contained <sup>3</sup>H that was nondetectable and below the RSRL.
- Southeast Transect: Most of these samples contained detectable <sup>3</sup>H concentrations above the RSRL.
- South Transect: Very high concentrations of <sup>3</sup>H were detected at the 10- to 16-m mark (6,700 pCi/mL) and at the 45- to 54-m mark (57.5 pCi/mL); however, concentrations of <sup>3</sup>H with distance markedly decrease to the RSRL level. The highest <sup>3</sup>H concentration in vegetation collected from this location corelates very well with the <sup>3</sup>H concentration in a soil sample collected from this same location

Table 1. Tritium Concentrations (pCi/mL)(Analytical Uncertainty) in Overstory Vegetation with Distance from the Area G Perimeter Fence.

_	Distance from the Perimeter Fence										
<b>Transect Direction</b>	10 to 16 m	45 to 54 m	90 to 145 m	127 to 181 m	172 to 254 m						
North	1.8 (0.22)*	0.63 (0.18)*	0.60 (0.16)*	0.82 (0.17)*	1.1 (0.18)*						
Northeast	0.98 (0.18)*	0.95 (0.18)*	0.26 (0.15)	0.39 (0.15)	0.28 (0.15)						
East	0.56 (0.15)*	0.30 (0.15)	0.25 (0.15)	-1.0 (0.18)	0.41 (0.15)						
Southeast	0.95 (0.18)*	0.45 (0.15)	0.59 (0.16)*	0.65 (0.16)*	0.60 (0.16)*						
South	6700 (500)*	58 (4.4)*	0.42 (0.15)	-0.08 (0.14)	0.01 (0.14)						
Southwest	0.31 (0.15)	0.37 (0.15)	0.45 (0.15)	0.27 (0.15)	0.13 (0.15)						
Northwest	1.3 (0.19)*	0.63 (0.16)*	0.64 (0.16)*	0.25 (0.15)	0.18 (0.15)						

<sup>\*</sup>Values are equal to or greater than three times the level of analytical uncertainty and higher than the RSRL of 0.50 pCi/mL.

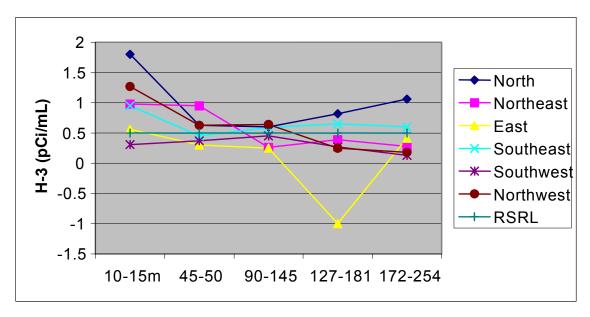


Figure 3. Trituin concentrations in trees as a function of distance and transect direction from the perimeter of Area G.

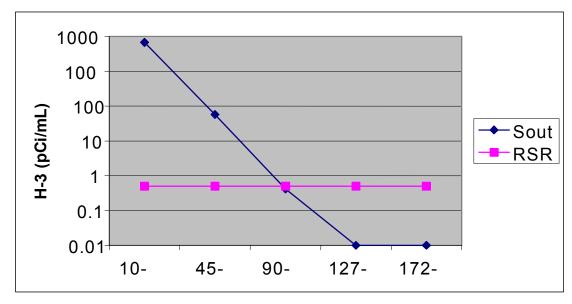


Figure 4. Tritium concentrations in trees as a function of distance and transect direction from the perimeter of Area G.

in 2003 (Nyhan et al., in preparation). That soil sample contained 22,000 pCi/mL of <sup>3</sup>H and exceeded soil screening action levels (6,600 pCi/mL). The high amount of <sup>3</sup>H in vegetation collected from the south transect (3.8E04 pCi/g-dry), however, was far below the Toxicity Reference Value (TRV) of 2E06 pCi/g-dry (Fresquez et al., 2003).

- Southwest Transect: All samples were nondetectable and within the RSRL.
- Northwest Transect: Concentrations of <sup>3</sup>H above the RSRL were detected to the 145-m mark; concentrations generally decrease to the RSRL thereafter.

#### IV. CONCLUSIONS

There were significantly higher concentrations of <sup>3</sup>H in overstory vegetation directly around the perimeter of Area G as compared to the regional background level and this agrees with past monitoring results. Two sides, the north and southeast transects, contained <sup>3</sup>H in the vegetation probably further than the distances sampled in this study. The highest concentration, 6,700 pCi/mL (3.8E04 pCi/g-dry), detected on the south side was correlated with past soil results; however, this amount was far below the TRV of 2E06 pCi/g-dry. For the most part, <sup>3</sup>H concentrations in overstory vegetation generally decreased with distance from the Area G fence perimeter.

#### V. ACKNOWLEDGMENTS

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#### APPENDIX A

## ACTUAL DISTANCES OF SAMPLE SITES WITHIN EACH TRANSECT (METERS FROM PERIMETER FENCE LINE AT AREA G)

North	16	50	110	150	205
Northeast	15	50	100	160	200
East	10	54	100	145	173
Southeast	10	50	100	150	200
South	10	50	145	181	254
Southwest	10	45	90	127	172
Northwest	10	45	100	145	200

#### APPENDIX B

## ANALYTICAL REPORTS OF TRITIUM IN UNWASHED OVERSTORY VEGETATION SAMPLES AT AREA G IN 2003

#### Sample Results Summary

Client Name: ESH20\_LANL

Client Project Name: Area-G Tritium Migration

Client Project Number: 7H05 WE6G

Laboratory Name: Paragon Analytics, Inc.

PAI Work Order: 0306090

Page: 1 of 4

Reported on: Wednesday, July 09, 2003

08:46:52

Lab Sample ID	Client Sample ID	Test	Nuclide	Result +	<i>l</i> - 2	s TPU	MDC	Units	Matrix	Prep Batch	Date Analyzed	Flags
0306090-1	G-NW-10	TRITIUM	H-3	1.27	+/-	0.38	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-2	G-NW-45	TRITIUM	H-3	0.63	+/-	0.32	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-3	G-NW-100	TRITIUM	H-3	0.64	+/-	0.32	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-4	G-NW-145	TRITIUM	H-3	0.25	+/-	0.30	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-5	G-NW-200	TRITIUM	H-3	0.18	+/-	0.29	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-6	G-N-14.5	TRITIUM	H-3	1.80	+/-	0.43	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-7	G-N-50	TRITIUM	H-3	0.63	+/-	0.35	0.54	pCi/mL	Liquid	LS01716	7/5/03	
0306090-8	G-N-110	TRITIUM	H-3	0.60	+/-	0.32	0.48	pCi/mL	Liquid	LS01716	7/5/03	LT
0306090-9	G-N-150	TRITIUM	H-3	0.82	+/-	0.33	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-10	G-N-205	TRITIUM	H-3	1.06	+/-	0.36	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-11	G-NE-15	TRITIUM	H-3	0.98	+/-	0.35	0.48	pCi/mL	Liquid	LS01716	7/5/03	

#### Comments:

#### Data Package ID: H3W0306090-1

#### Qualifiers/Flags:

U - Result is less than the sample specific MDC.

LT - Result is less than Requested MDC, greater than sample specific MDC.

Y1 - Chemical Yield is in control at 100-110%. Quantitative Yield is assumed.

Y2 > Chemical Yield outside default limits.

Paragon Analytics Inc.

Abbreviations:

TPU - Total Propagated Uncertainty (see PAI SOP 743) MDC - Minimum Detectable Concentration (see PAI SOP 709)

#### Sample Results Summary

Client Name: ESH20\_LANL

Laboratory Name: Paragon Analytics, Inc.

Page: 2 of 4

Client Project Name: Area-G Tritium Migration

Client Project Number: 7H05 WE6G

PAI Work Order: 0306090

Reported on: Wednesday, July 09, 2003

08:46:52

Lab Sample ID	Client Sample ID	Test	Nuclide	Result +/-	2 s TPU	MDC	Units	Matrix	Prep Batch	Date Analyzed	Flags
0306090-12	G-NE-50	TRITIUM	H-3	0.95	+/- 0.35	0.48	pCi/mL	Liquid	LS01716	7/5/03	
0306090-13	G-NE-100	TRITIUM	H-3	0.26	+/- 0.30	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-14	G-NE-160	TRITIUM	H-3	0.39	+/- 0.30	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-15	G-NE-200	TRITIUM	H-3	0.28	+/- 0.30	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-16	G-W-10	TRITIUM	H-3	0.31	+/- 0.30	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-17	G-W-50	TRITIUM	H-3	0.14	+/- 0.29	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-18	G-W-100	TRITIUM	H-3	0.52	+/- 0.31	0.48	pCi/mL	Liquid	LS01716	7/5/03	LT
0306090-19	G-W-150	TRITIUM	H-3	0.24	+/- 0.29	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-20	G-W-200	TRITIUM	H-3	0.21	+/- 0.29	0.48	pCi/mL	Liquid	LS01716	7/5/03	U
0306090-21	G-E-10	TRITIUM	H-3	0.56	+/- 0.31	0.48	pCi/mL	Liquid	LS01717	7/6/03	LT
0306090-22	G-E-50	TRITIUM	H-3	0.30	+/- 0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U

#### Comments:

#### Data Package ID: H3W0306090-1

Qualifiers/Flags:

U - Result is less than the sample specific MDC.

LT - Result is less than Requested MDC, greater than sample specific MDC.

Y1 - Chemical Yield is in control at 100-110%. Quantitative Yield is assumed.

Y2 - Chemical Yield outside default limits.

TPU - Total Propagated Uncertainty (see PAI SOP 743) MDC - Minimum Detectable Concentration (see PAI SOP 709)

Paragon Analytics Inc.

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### Sample Results Summary

Client Name: ESH20\_LANL

Client Project Number: 7H05 WE6G

Laboratory Name: Paragon Analytics, Inc.

Page: 3 of 4

Client Project Name: Area-G Tritium Migration

PAI Work Order: 0306090

Reported on: Wednesday, July 09, 2003

08:46:53

Lab Sample ID	Client Sample ID	Test	Nuclide	Result +/-	2 s TPU	MDC	Units	Matrix	Prep Batch	Date Analyzed	Flags
0306090-23	G-E-100	TRITIUM	H-3	0.25 +	/- 0.29	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-24	G-E-150	TRITIUM	H-3	-1.00 +	/- 0.36	0.60	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-25	G-E-200	TRITIUM	H-3	0.41 +	/- 0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-26	G-SE-10	TRITIUM	H-3	0.95 +	/- 0.35	0.48	pCi/mL	Liquid	LS01717	7/6/03	
0306090-27	G-SE-50	TRITIUM	H-3	0.45 +	/- 0.31	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-28	G-SE-105	TRITIUM	H-3	0.59 +	/- 0.32	0.48	pCi/mL	Liquid	LS01717	7/6/03	LT
0306090-29	G-SE-150	TRITIUM	H-3	0.65 +	/- 0.32	0.48	pCi/mL	Liquid	LS01717	7/6/03	
0306090-30	G-SE-200	TRITIUM	H-3	0.60 +	/- 0.32	0.48	pCi/mL	Liquid	LS01717	7/6/03	LT
0306090-31	G-S-10	TRITIUM	H-3	6700 +	/- 1000	2.1	pCi/mL	Liquid	LS01717	7/6/03	
0306090-32	G-S-50	TRITIUM	H-3	57.5 +	/- 8.8	0.54	pCi/mL	Liquid	LS01717	7/6/03	
0306090-33	G-S-100	TRITIUM	H-3	0.42 +	/- 0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U

#### Comments:

#### Data Package ID: H3W0306090-1

#### Qualifiers/Flags:

U - Result is less than the sample specific MDC.

LT - Result is less than Requested MDC, greater than sample specific MDC.

Y1 - Chemical Yield is in control at 100-110%. Quantitative Yield is assumed.

Abbreviations:

TPU - Total Propagated Uncertainty (see PAI SOP 743)

MDC - Minimum Detectable Concentration (see PAI SOP 709)

Paragon Analytics Inc.

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### Sample Results Summary

Client Name: ESH20\_LANL

Client Project Number: 7H05 WE6G

Laboratory Name: Paragon Analytics, Inc.

Page: 4 of 4

Client Project Name: Area-G Tritium Migration

PAI Work Order: 0306090

Reported on: Wednesday, July 09, 2003

08:46:54

Lab Sample ID	Client Sample ID	Test	Nuclide	Result +	·/- 2	s TPU	MDC	Units	Matrix	Prep Batch	Date Analyzed	Flags
0306090-34	G-S-150	TRITIUM	H-3	-0.08	+/-	0.28	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-35	G-S-200	TRITIUM	H-3	0.01	+/-	0.28	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-36	G-SW-10	TRITIUM	H-3	0.31	+/-	0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-37	G-SW-50	TRITIUM	H-3	0.37	+/-	0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-38	G-SW-100	TRITIUM	H-3	0.45	+/-	0.31	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-39	G-SW-150	TRITIUM	H-3	0.27	+/-	0.30	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-40	G-SW-200	TRITIUM	H-3	0.13	+/-	0.29	0.48	pCi/mL	Liquid	LS01717	7/6/03	U
0306090-41	G-JEMEZ	TRITIUM	H-3	-0.08	+/-	0.28	0.48	pCi/mL	Liquid	LS01719	7/6/03	U
0306090-42	G-WATER	TRITIUM	H-3	-0.08	+/-	0.28	0.48	pCi/mL	Liquid	LS01719	7/6/03	U

#### Comments:

Data Package ID: H3W0306090-1

Qualifiers/Flags:

U - Result is less than the sample specific MDC.

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